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Safeguards Science and Technology
Group (NEN-1)

Nuclear Nonproliferation Division

COE1 Calorimeter

Operations Manual

November, 2015

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Applicability:

This document applies to the usage of the COE1 calorimeter to perform measurements on Pu bearing items.

History of Revisions

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1. Overview

1.1. Purpose

The purpose of this manual is to describe the operations of the COE1 calorimeter which is used to measure the thermal power generated by the radioactive decay of plutonium bearing materials for the purposes of assaying the amount of plutonium within the material.

1.2. Audience

The intended audiences for this manual are the operators, instructors, and managers of the China Center of Excellence who are responsible for operating and using the COE1 calorimeter.

1.3. Acronyms

COE — China Center of Excellence

DOE — Department of Energy

EHS — Electrical Heat Standard

GPIB — General Purpose Interface Bus

LANL — Los Alamos National Laboratory

NDA — Nondestructive Assay

NEN-1 — Safeguards Science and Technology Group, Los Alamos National Laboratory

SNM — Special Nuclear Material

1.4. Document Overview

Section 2 contains a description of the main components of the COE1 calorimeter

Section 3 contains a description of the ancillary components of the COE1 calorimeter

Section 4 contains the contact information for additional technical support.

2. COE1 Calorimeter Overview

This section provides an overview of the COE1 calorimeter including its construction, mechanical and electrical components, and calibration.

2.1. General

The COE1 calorimeter is an instrument that measures the thermal power generated by the decay of radioactive materials. The system is to be used within the China Center of Excellence (COE) as a training tool to teach individuals how to perform calorimetric assay. The system can perform accurate measurements of any solid nuclear material with a thermal power greater than 0.06 Watts. This includes material containing plutonium or ^{241}Am . Material containing enriched uranium cannot be measured with this system because its thermal power is too low. **The measurement of items with a thermal power greater than 20 Watts is not recommended due to the potential for overheating the measurement chamber.**

The thermal-power measurement system consists of (1) a twin-bridge calorimeter that is surrounded by a temperature-controlled water bath and located inside an insulated 110-gal. stainless-steel drum, (2) an instrument rack containing the multimeters that are used to measure the calorimeter and environmental conditions as well as a printer and, and (3) a PC used for data acquisition that is connected to the multimeters using an IEEE-488 General Purpose Interface Bus (GPIB). A thermoelectric unit attached to the calorimeter provides heating and cooling to maintain a constant temperature environment for the calorimeter. The calorimeter system is designed to run continuously (24 hours a day/7 days a week) powered by 120 V AC, even when measurements are not being taken.

The calorimeter, electronics cabinet, water bath supply, and temperature control are shown in Figure 1. This calorimeter was built at Mound Laboratory at Miamisburg, Ohio, USA in 1986, and shipped to Los Alamos in 1996. The calorimeter was reactivated in 2011. The calorimeter measurement chamber is 7" diameter x 12.5" tall. It was designed to hold items as large as a one-gallon plastic container.



Figure 1. COE1 Calorimeter System

A calorimeter measurement consists of placing an item in the measurement chamber, waiting until the temperature of the object has reached thermal equilibrium, and recording its thermal power from the data acquisition computer using MultiCal software¹. For most items the measurement time can range from 5 - 12 hours depending on the initial temperature, heat conductivity, mass, specific heat of the sample, and whether the calorimeter is operated in passive or servo mode. The calorimeter measurement chamber from the COE1 calorimeter is shown in Figure 2 for reference. The temperature sensing assembly is called the 'thermel'

¹ Operation of MultiCal is described in "MultiCal Version 4.0 Users Manual, LA-UR-03-8204, February 2004

(Thermal element). After the item is placed in the chamber an insulating plug, shown in Figure 3, is inserted to thermally isolate the chamber from the room temperature environment. A layer of thermal insulation is permanently located under the measurement chamber. The measurement chamber is made of aluminum around which is wound electrical heater wire forming part of the thermel. This allows electrical thermal power to be introduced into the calorimeter from an external power supply.

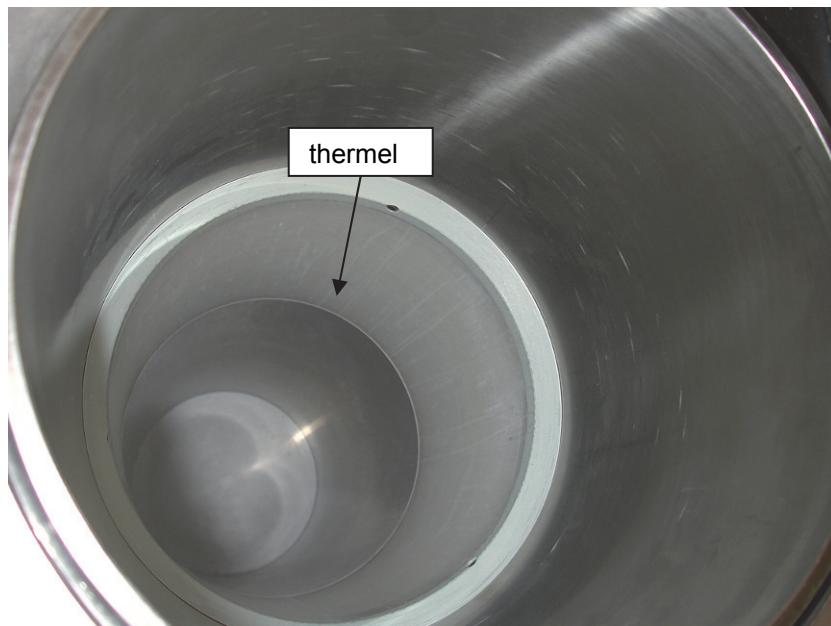


Figure 2. Calorimeter measurement well.

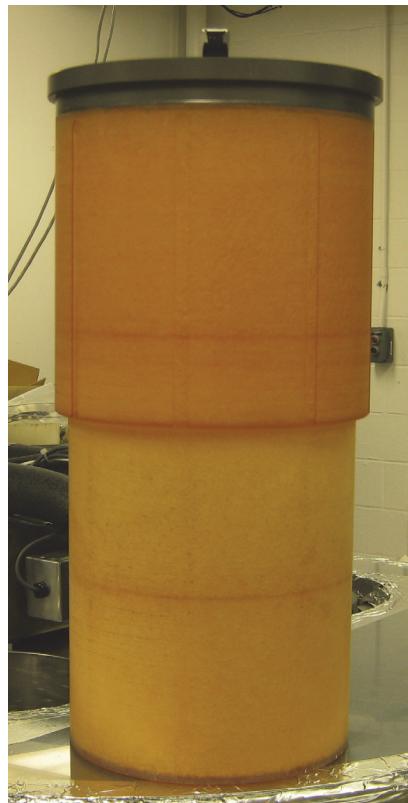


Figure 3. Insulating Plug

Surrounding the measurement thermel is a stainless steel jacket that separates the thermel and measurement chamber from the water bath. Figure 4 shows the stainless steel jackets for the measurement thermel and the reference thermel. The outside of the thermel is partially insulated from the inside of the stainless steel water jacket by a thin layer of air. Due to this partial insulation the heat from the radioactive material raises the temperature of the measurement chamber and its contents above the temperature of the water bath. The difference in temperature between the measurement chamber outer wall and the water-bath jacket inner wall is measured using long lengths of nickel (Ni) wire. The resistance of the Ni wire sensor material increases $\sim 0.6\%/\text{ }^{\circ}\text{C}$. Thus the resistance of these wires increases with the increased temperature due to the radioactivity. A duplicate thermel and stainless steel jacket is located adjacent to the measurement jacket as shown in Figure 4. The reference thermel is wound identically with two lengths of Ni wire as the measurement thermal and is maintained at the water bath temperature. The response of the two thermels to temperature drifts and fluctuations of the water is nearly identical and cancels out much of this measurement noise.



Figure 4. Calorimeter water jacket.

2.2. Electrical wiring

The four sensor wires are wound in a Wheatstone Bridge configuration, as shown in the right side of Figure 5. A constant current source (10 milliAmps) is used to generate voltages across the arms of the Wheatstone Bridge. The change in resistances R_{S1} and R_{S2} due to the temperature rise caused by the heat versus the constant reference resistances (R_{R1} and R_{R2}) held at a constant temperature is transformed into a change in voltage across the arms of the Bridge, a change in the bridge potential (BP). The electrical circuit associated with the bridge is shown in the middle of Figure 5. The change in voltage is measured with an accurate digital voltmeter (Keithley Model 2001). With no sample in the chamber the bridge potential is called the baseline. The four bridge arm resistances are not exactly equal so the baseline is not equal to zero Volts. Additional electrical meters shown in Figure 5 are used to monitor the electrical current source, bath temperature, and room temperature since large variations in these components could affect the quality of the measurement.

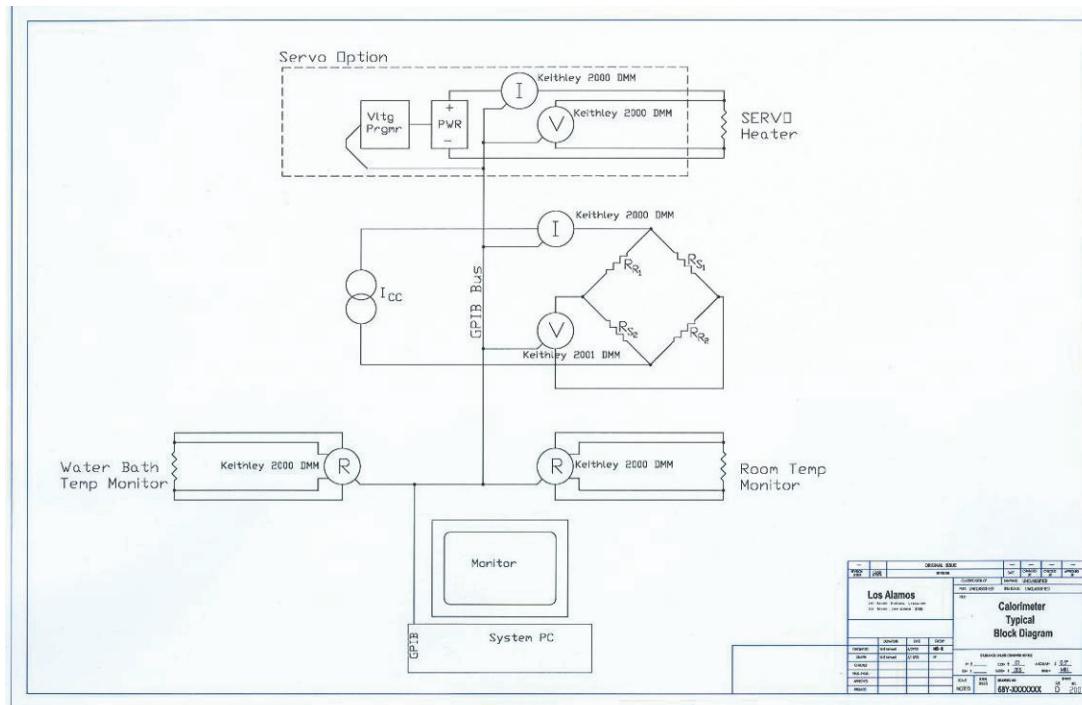


Figure 5. Calorimeter electrical schematic.

2.3. Calibration

After a sample is loaded in the calorimeter the increase of BP above the baseline approaches an equilibrium value with a magnitude proportional to the thermal power of the sample. The change in bridge potential with thermal power, the sensitivity of the calorimeter, is 0.01376 Volts/Watt. This calibration factor is obtained by generating known amounts of power in the measurement chamber using radioactive heat standards or using a calibrated electrical resistance heater and measuring the changes in BP at different power levels. The change in BP is nearly linear with increasing power.

Calibration data for the calorimeter are shown in Figure 6 along with a 2nd order least squares fit to the data. This calibration curve is valid for a water bath temperature of 25 °C. The calibration data were obtained by measuring calibrated encapsulated Pu-238 oxide heat standards. The ²³⁸Pu heat standards themselves had been calibrated against electrical standards traceable to the National Institute of Standards and Technology (NIST). The calibration curve has a slight decrease in sensitivity with increasing power, typical for this type of calorimeter. The instrument has been calibrated for items ranging from 0.07 to 10 Watts thermal power. Measurement of items outside of this range may require re-calibration.

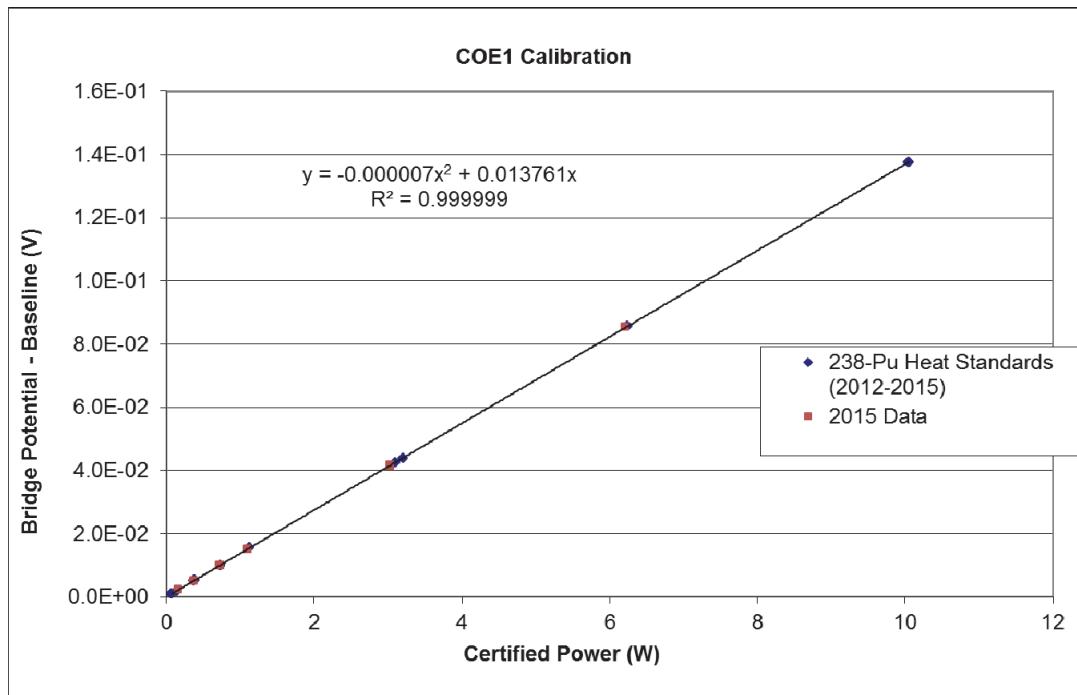


Figure 6. Calibration curve for COE1 calorimeter. The data used to create the calibration curve is represented by the blue diamonds while the red squares show measurements used to test the calibration curve.

Table 1 shows the summary of the COE1 calorimeter measurements that were made with certified ^{238}Pu heat standards at LANL prior to shipment of the calorimeter to the COE.

Table 1 Calibration Data using ^{238}Pu Heat Standards for COE1

Date	Standard ID	Delta BP (Volts)	Standard Power (Watts)
6/5/2012	0.1WB	0.0009714	0.071384
6/7/2012	MAD-1469-F79	0.0424973	3.100831
6/8/2012	0.2WC	0.0018810	0.137598
6/26/2012	1.5WJ	0.0155314	1.129581
6/28/2012	0.5WI	0.0052558	0.381859
7/3/2012	0.1WB	0.0009737	0.071341
7/17/2012	0.5WI	0.0052320	0.381703
7/18/2012	1.5WJ	0.0154915	1.129047
7/19/2012	0.1WB	0.0009821	0.071316
7/20/2012	0.2WC	0.0018767	0.137473
7/27/2012	0.1WB	0.0009580	0.071304
8/3/2012	MAD-149-F79	0.0425261	3.097031
8/6/2012	1.5WJ	0.0155560	1.128585
8/7/2012	0.5WI	0.0052301	0.381531
6/25/2014	1.0WJ	0.0101719	0.730496
9/30/2014	UAS-1403-B85	0.0439876	3.200073
10/6/2014	0.5WI	0.0051596	0.375103
10/8/2014	UAS-1403-B85+MAD-1469-F79	0.0856467	6.243976
10/15/2014	0.1WB	0.0009504	0.070072
10/20/2014	1.0WJ	0.0100309	0.728661
10/22/2014	UAS-1403-B85	0.0439938	3.198557
10/29/2014	0.1WB	0.0009644	0.070051
11/4/2014	0.5WI	0.0052226	0.374869
11/17/2014	UAS-1403-B85+MAD-1469-F79	0.0856574	6.238602
11/19/2014	1.0WJ	0.0100366	0.728191
5/26/2015	Stack of seven stds*	0.1375100	10.049062
6/2/2015	Stack of seven stds*	0.1376018	10.047549
6/9/2015	Stack of seven stds*	0.1376124	10.046034

*Stack of seven stds – UAS-1403-B85, MAD-1469-F79, 1.5WG, 1.5WJ, 1.5WF, 0.5WI, and 0.2WC

2.4. Modes of Operation

Two modes of operation are possible with this calorimeter, passive and servo. In the passive mode the power of the item is measured by measuring the change in bridge potential. The calibration curve is used to obtain the item power. In the servo mode of operation, also called the power replacement mode, the built-in heater is used to deliver a constant amount of power into the measurement chamber. The bridge potential generated by this introduction of heat is used to control the power supply that is generating the current passing through the 100-ohm resistance heater in the calorimeter heater form. In servo mode, an increase or decrease in the measured Bridge Potential from a given set point causes a signal to be sent to the power supply controller to increase or decrease the delivered power from the power supply to maintain the set point BP. The amount of power that is supplied with no sample in the calorimeter is referred to as the basepower. When a radioactive sample is introduced the radioactive heat replaces a portion of electrical heat generated by the power supply. The amount of power supplied by the power supply is decreased by the amount of power generated by the sample. The shift in this externally supplied power equals the sample power. Calorimeter measurement times are shorter in the servo mode of operation compared to the passive mode. Passive mode measurements are more accurate. In Figure 5 two multimeters are used to measure the current and voltage across the calorimeter heater generated by the power supply. The product of these values is used to calculate the electrical power injected into the calorimeter.

In either passive or servo mode, the bias is expected to be small. The relative precision of the calorimeter is the worst at the lowest power. For passive mode, the replicate ^{238}Pu standard measurements listed in Table 1 provide estimates of the precision for different powers. Listed in Table 2 is the precision and bias of COE1 that were achieved at LANL at different power levels.

Table 2 Precision Estimates for COE1.

Power (W)	% precision (RSD)	% bias (RSD)
0.07	0.93	-0.93
0.38	0.61	0.08
0.73	0.35	0.08
1.1	0.19	-0.03
3.1	0.15	-0.03
6.2	0.06	0.07
10.0	0.02	0.04

The bias and precision of the COE1 calorimeter can be impacted by a number of different conditions associated with the operations of the calorimeter. These include

- 1) Variations in the room temperature which may affect the stability of the reference temperature.
- 2) Extreme mechanical shock (e.g. dropping the calorimeter or hitting the calorimeter with a large object)
- 3) Moving the instrument to another location within the room or to a different room.
- 4) A change in the bath temperature setting.
- 5) Damage to the inside of the calorimeter during insertion or removal of the calorimeter can.
- 6) Noise generated in the electronics due to an external AC power supply.

In the case where the bath temperature setting is changed, the calibration of the calorimeter would have to be reevaluated as changing the reference temperature associated with the calorimeter can impact that response of the calorimeter. For the other conditions listed above, measurements of known standards are recommended to check that the performance of the calorimeter is the same as prior to the event occurring.

3. Ancillary Calorimeter System Components

This section describes the ancillary components of the COE1 calorimeter including the water bath, calorimeter can, electronics, and the electrical heat standard.

3.1. Water Bath

The water bath provides a constant temperature environment for the calorimeter. The water bath consists of a 110-gallon stainless steel drum that is shown in Figure 7. This drum is surrounded by a layer of reflective insulation and a layer of stainless steel. The constant temperature water is maintained by a thermoelectric heat exchanger attached to the side of the drum. Water is pumped past the heat exchanger, which provides Peltier cooling or resistance heating depending on the set point temperature on the Hart Model 2100 Controller unit attached adjacent to the thermoelectric unit. The control temperature is set on the Hart controller at 25 °C. A thermistor probe inserted into the bath monitors the bath temperature for the controller unit. This sensor is inserted into the top of the water bath tank. Another thermistor sensor is inserted into the water bath to monitor the water bath temperature attached to an ohmmeter (Keithley 2000) connected to the data acquisition network. The

water bath is replenished infrequently through a feed container. The use of distilled or deionized water is recommended to prevent a build up of scale in the calorimeter water jacket.



Figure 7. 110 gallon water bath. The circles indicate the fill and drain ports for the drum.

3.2. Calorimeter can

Items to be measured are nominally placed in a calorimeter can shown in Figure 8 in order to minimize the possibility of contaminating the sample chamber of the calorimeter. The item to be measured is typically placed in the middle of the can. This can is machined to closely fit inside the measurement chamber. After placing the item in the can the empty space in the can may be filled with low specific heat and high thermal conductivity material such as Al foil. The addition of Al foil decreases the measurement time and reduces thermal noise in the system. Once the cal can lid is attached to the cal can, the cal can is carefully lowered into the measurement well. For items which can fit in the calorimeter measurement chamber, but cannot fit into the cal can, the item can be directly placed the measurement chamber provided that Al foil is used to ensure good contact between the chamber walls and the item.



Figure 8. COE1 calorimeter can.

3.3. Electronics

Figure 9 shows the electronics used to operate and measure the data from the calorimeter. The electrical connections between the sensors and meters and the data acquisition computer are shown in Figure 5. The electronics consist of Keithley Model 2000 and 2001 multimeters which are capable of accurate voltage and current measurements and are used to measure bridge potential, bridge current and the resistances of the thermistor probes. The constant current source used to excite the bridge is an InstruLab Even-Volt DC power supply that generates 10 milliAmps of current. The data from the meters are transmitted using a General Purpose Interface Bus (GPIB) to the Dell Laptop which runs the MultiCal 4.0.22 data acquisition software. For operating the calorimeter in servo mode, a KEPCO Model JQE 0 - 25 V, 0 - 4 Amp power supply is used to provide heating to the calorimeter. The power supply is controlled by a KEPCO Programmer Model SN 488-032. The electrical connections from the meters to the sensors are made to the back of the meters and are shown in Figure 10.



Figure 9. Electronics used for calorimeter operation. Each multimeter in the cabinet is labeled indicating its purpose.

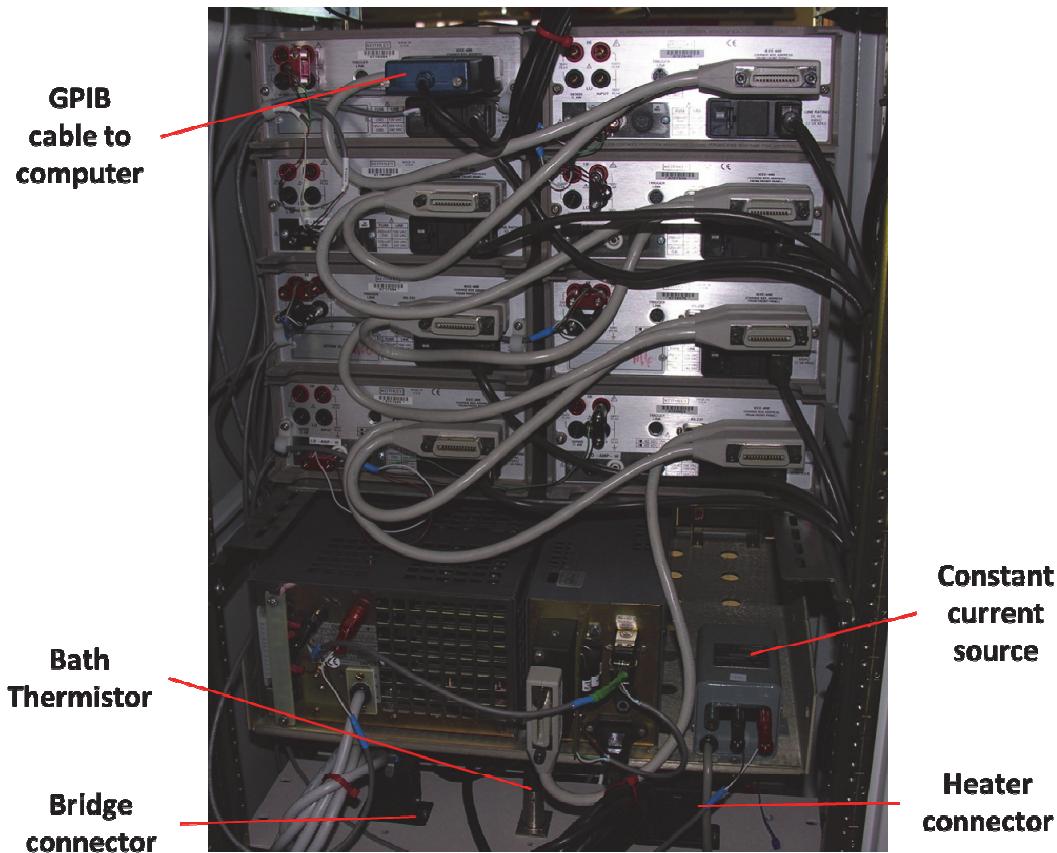


Figure 10. Electrical connections for the COE1 calorimeter.

3.4. Electrical Heat Standard

A power supply that serves as an electrical heat standard (EHS), supplying known amounts of power to the built-in calorimeter heater, is shown in Figure 11. A cable from this box plugs directly into the calorimeter built-in heater through the calorimeter 8-pin connector that is also used for the servo mode of operation. The EHS is only be used for passive mode of operation. The power range of the EHS is 0.01 to 10 Watts. The EHS output has been calibrated using Pu-238 heat standards as shown in Table 3. Figure 12 shows the calibration of the EHS as a function of the measured calorimeter power while Table 4 shows the results of the electrical heat standards calibration.

To ensure that the power output from the EHS remains constant over time, the monitor output from the EHS is connected to two Keithley multimeters in the electronics rack and recorded in MultiCal as a separate device labeled 'EHS'. The output from the monitor is directly proportional to the power output of the EHS as shown in Figure 13. Based on the data shown in Figure 13, a scaling factor of 1.7032 is needed to convert the monitor output to the actual power output of the EHS. Table 5 presents the data associated with the monitor output and the EHS power setting.

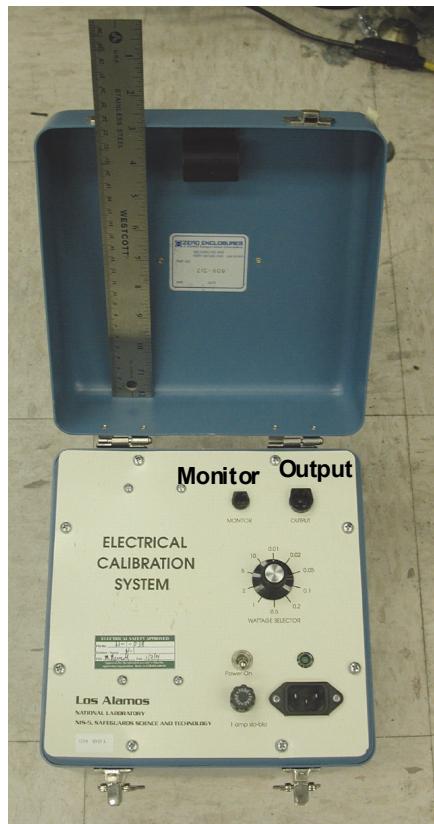


Figure 11. Electrical Heat Standard.

Table 3 Calibration of COE1 Electrical Heat Standard

Date	Electrical Heat Standard Setting (Watts)	Actual Power (Watts)	Monitor Output (Watts)
7/10/2015	1	0.98046	0.57751
7/15/2015	1	0.98022	0.57725
7/22/2015	1	0.98130	0.57716
7/23/2015	0.1	0.09744	0.05729
7/27/2015	5	4.91367	2.88408
7/30/2015	0.1	0.09803	
8/3/2015	5	4.91552	
8/5/2015	0.5	0.49033	
8/7/2015	1	0.98013	
8/14/2015	1	0.97993	
8/18/2015	10	9.85319	
8/20/2015	5	4.91504	
8/24/2015	0.1	0.09845	0.05731
8/26/2015	0.5	0.48952	0.28760
8/28/2015	1	0.98095	0.57661
9/1/2015	10	9.85051	5.78325
9/3/2015	5	4.91615	2.88597
9/8/2015	5	4.91652	2.88520
9/10/2015	0.5	0.48975	0.28762
9/17/2015	2	1.95670	1.15029
9/21/2015	0.1	0.10245	0.05732
10/1/2015	5	4.91414	2.88327
10/5/2015	10	9.86075	5.79117
10/7/2015	1	0.98207	0.57708
10/20/2015	10	9.86036	5.79101

Table 4. COE1 EHS Calibration.

Setting (Watts)	Calibrated Power (Watts)	Std Dev (Watts)
0.1	0.09909	0.00228
0.5	0.48986	0.00041
1	0.98072	0.00077
5	4.91517	0.00112
10	9.85482	0.00531

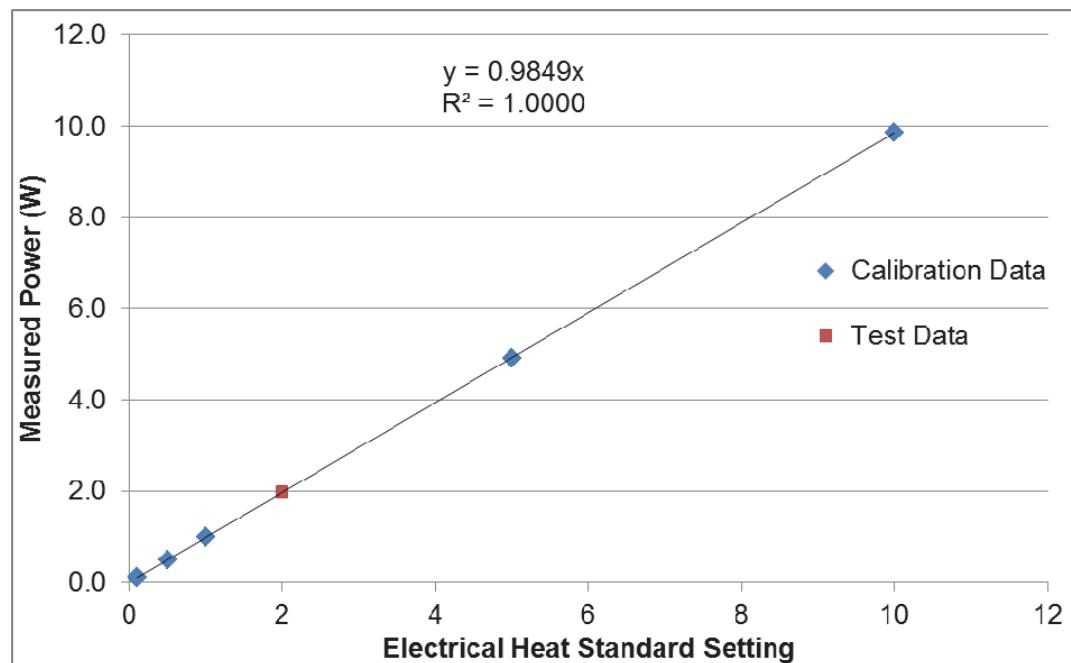


Figure 12 Measured thermal power as a function of the Electrical Heat Standard dial setting for COE1.

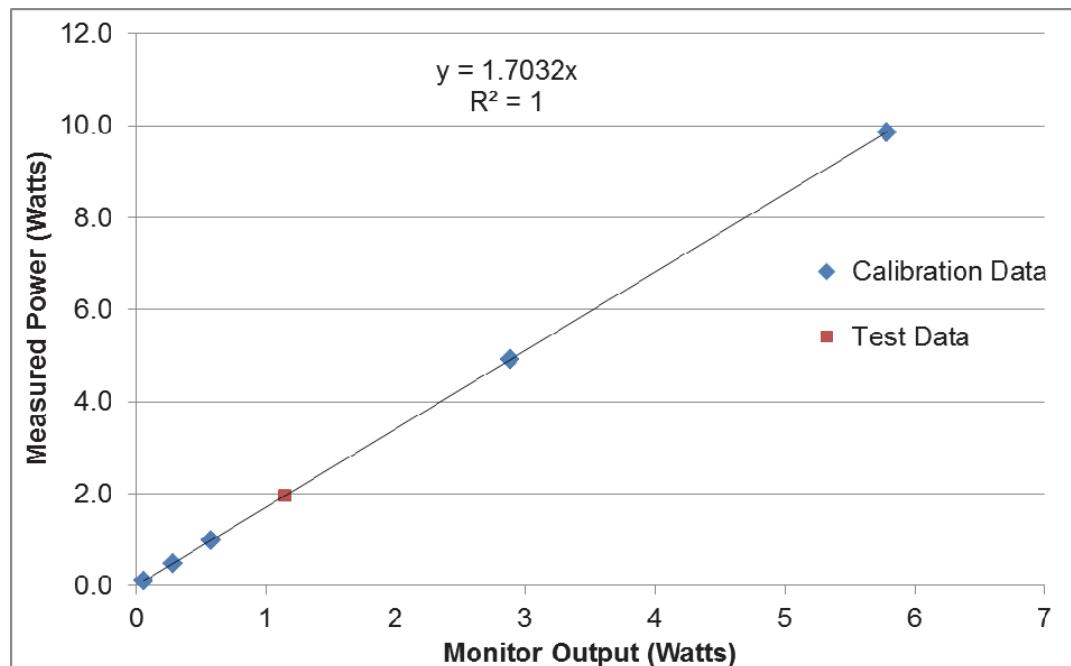


Figure 13. EHS Power output as a function of the monitor output.

Table 5. EHS Monitor Output

Setting (Watts)	Monitor Output (Watts)	Std Dev (Watts)
0.1	0.05731	0.00001
0.5	0.48994	
1	0.57712	0.00033
5	2.88463	0.00120
10	5.78721	

4. Contact Information

For further assistance, contact the following LANL NEN-1 personnel:

Mark Croce, (505) 665-2635, mpcroce@lanl.gov

Safeguards Science and Technology Group (NEN-1), (505) 667-7110